

## LONG-TERM OBSERVED OZONE TRENDS IN THE FREE TROPOSPHERE AND LOWER STRATOSPHERE

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### ABSTRACT

The vertical distributions of ozone trends in the free troposphere and lower stratosphere were derived from ozonesonde observations taken over an average period of  $\sim 20$  years. The results for the annual trends show a consistent pattern of increased ozone of  $\sim 1\%/yr$  to  $2\%/yr$  up to  $\sim 300$  mb and decreased ozone of  $\sim -0.6\%/yr$  from  $\sim 100$  to  $50$  mb.

Statistically significant positive trends found in mid-troposphere ( $\sim 500$  mb) at a set of representative stations in the Northern Hemisphere have little apparent seasonal variation. Negative trends are generally strongest at  $50$ – $70$  mb with a tendency to be larger during spring. A highly significant negative trend of  $\sim -5\%/yr$  is found near  $100$  mb over Syowa ( $69^\circ S$ ) during spring.

### INTRODUCTION

Documentation of the three-dimensional distribution of atmospheric ozone in the troposphere and lower to middle stratosphere and its time variations have been matters of some concern because of, among other things, its dominant role in regulating the UVB irradiance at the ground and its potential role in problems of climatic change (WMO, 1989; IPCC, 1990).

Direct measurements of the ozone concentration up to  $\sim 30$  km have been made routinely with the aid of balloon-borne instruments since about 1962 (see, for instance, London and Liu, 1992). It has been shown from data derived from these ozonesonde measurements in the global observing network that during the past 20 or so years there has been a statistically significant negative annual ozone trend in the lower stratosphere and a positive trend in the mid-troposphere (e.g., WMO, 1989). Here, we discuss the annual and seasonal variations of these trends as observed at a few stations with relatively long periods of observations. The types of instruments used at these stations and the time periods of the observations up to 1988 are given in London and Liu (1992). In this paper the data set is generally extended to 1990.

The results from three different groups of stations are discussed below. These results are based on observations from two stations in Central Europe (Hohenpeissenberg and Payerne), a station in the Antarctic (Syowa), and a composite from four stations in Canada. In each case the annual and seasonal trends have been analyzed except for Syowa, where data were sufficiently available for analysis only for the spring season (i.e., September, October, and November).

### DATA ANALYSIS

For the annual trends all observations for each month were averaged first, and then the 12 monthly values were averaged for each year. For the four quarterly trends all observations for each quarter were averaged separately for each year. Quarter 1 covered December, January, and February; quarter 2 covered March, April, and May, etc. For the four Canadian stations we assumed that the observed data were taken from a common population; therefore, all observations for monthly or quarterly periods were grouped together as a Canadian composite. The four Canadian stations for which there were relatively long periods of ozonesonde observations are Resolute ( $75^\circ N$ , 1966–1990), Churchill ( $59^\circ N$ , 1974–1990), Edmonton ( $53^\circ N$ , 1973–1990), and Goose ( $53^\circ N$ , 1970–1990). The starting period for each of the Canadian stations was different. The composite analysis period started in 1973, the year when three of the four stations were operative.

Ozonesonde observations from two representative European stations with long periods of available data sets were used. These stations are Hohenpeissenberg ( $48^\circ N$ , 1967–1990) and Payerne ( $47^\circ N$ , 1969–1988).

The only station in the Southern Hemisphere for which there is an available quasi-continuous set of measurements is Syowa ( $69^\circ S$ , 1966–1990), and even here the observing series is adequate for analysis only during the spring (September, October, and November) season. Trends were calculated for 11 standard levels for which ozonesonde data are available from the World Ozone Data Center, Atmospheric Environment Service, Canada. Values from Payerne were interpo-

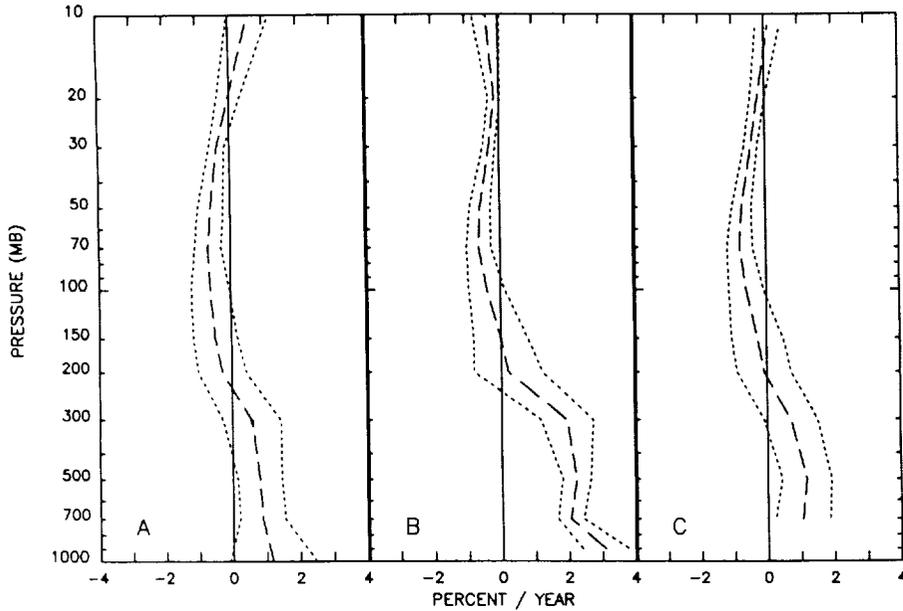


Fig. 1. Annual ozone trends.  
 (A) Canadian composite, 1973-90.  
 (B) Hohenpeissenberg, 1967-90.  
 (C) Payerne, 1969-88.

lated to these levels from the mean layer data supplied to us by J. Staehelin.

## RESULTS

### Annual Trends

The annual ozone trends (dashed lines) are given in Fig. 1 for the Canadian composite (1973-1990), Hohenpeissenberg (1967-1990), and Payerne (1969-1988). The dotted lines represent the standard error ( $\pm 2\sigma$ ) of the individual annual trend values for the periods as indicated. The vertical distributions of the annual trends for this representative set are very similar to other pub-

lished results, generally for shorter periods (e.g., Tiao et al., 1986; Wege et al., 1989; Staehelin and Schmid, 1991; Stolarski et al., 1992; McCormick et al., 1992).

Large, significant positive ozone trends of  $\sim 1\%/yr$  to  $2\%/yr$  are found in mid-troposphere (500-700 mb), and significant negative trends,  $-0.6\%/yr$  to  $-0.7\%/yr$ , are found at 50-70 mb. The crossover from positive to negative trends occurs at a pressure level of  $\sim 200$  mb, in the region of the diffuse tropopause layer at these latitudes. Inspection of the annual negative trends in the lower and middle stratosphere for each of the four Canadian stations (not shown here) indicates almost

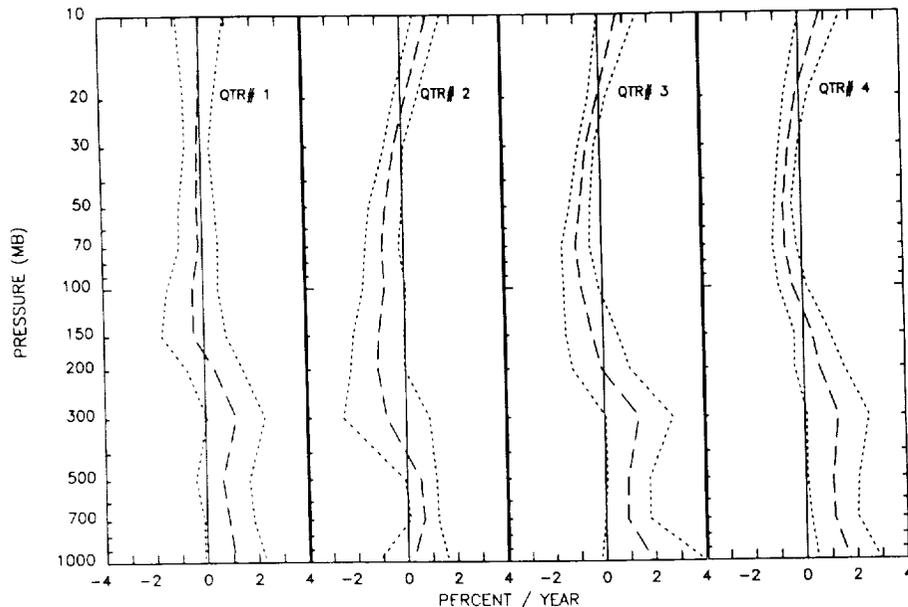
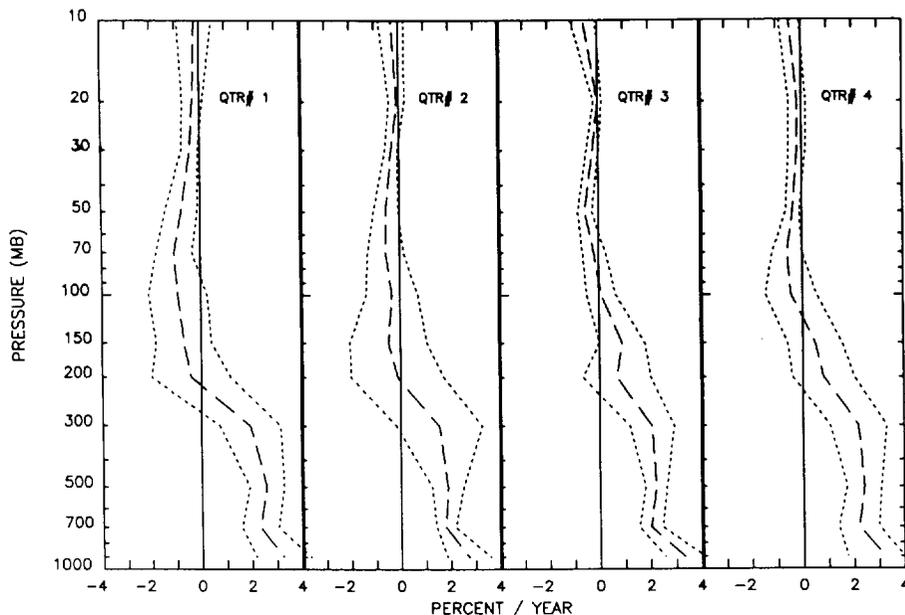


Fig. 2a. Ozone trends (quarterly), Canadian composite, 1973-90.

Fig. 2b. Quarterly ozone trends, Hohenpeissenberg (48°N), 1967-90.



the same values. However, the mid-tropospheric positive trends have some differences indicating year-to-year local influences, both chemical and dynamic, affecting the tropospheric trends. The largest standard errors associated with the trend values are generally found at the surface and in the upper troposphere at ~300 mb.

Seasonal Variations

The seasonal trends for the Canadian composite values are given in Fig. 2a. Although the ozone trends in the troposphere are positive (~1.0%/yr) during all seasons, the trends are just barely statistically significant.

There are, however, large significantly negative trends between 50 and 200 mb in spring and between 30 and 100 mb in summer and fall. Note that the ozone trends over Canada in winter (quarter 1) are close to zero. This is true for three of the four stations in the composite set. The crossover level from positive to negative trends varies with the seasons from ~175 mb during winter to 400 mb in spring and then to lower pressures (~125 mb) in fall similar to the seasonal shift of tropopause height at these latitudes.

The calculated seasonal trends for Hohenpeissenberg and Payerne (Figs. 2b, 2c) show very similar distri-

Fig. 2c. Quarterly ozone trends, Payerne (47°N), 1969-88.

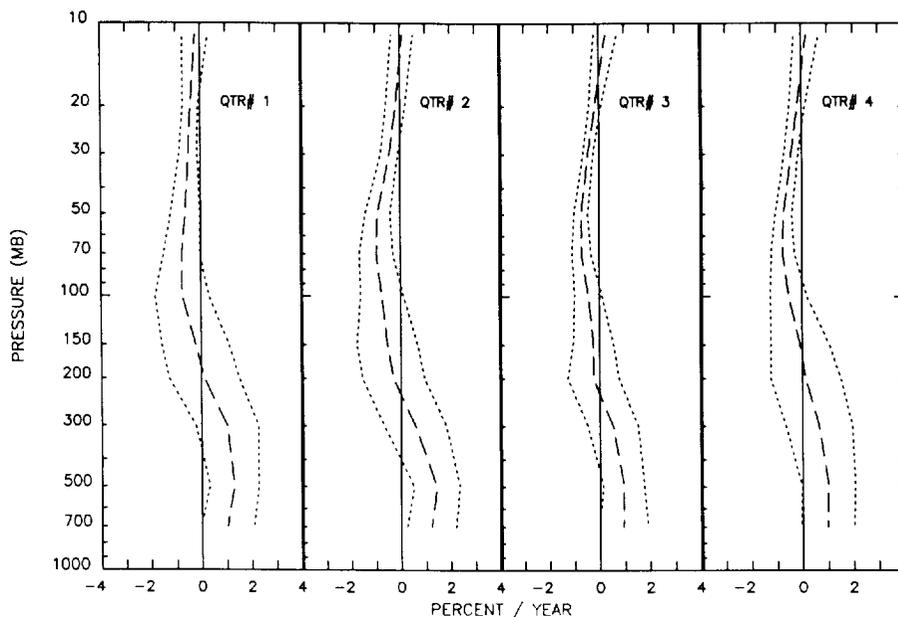


Table 1. Ozone Trends (% per year)

Level (mb)	Annual	Qtr. 1	Qtr. 2	Qtr. 3	Qtr. 4
<b>Canadian Composite (1973 - 1990)</b>					
Surface	1.19	1.10	0.21	1.89	1.70
700	0.88	0.88	0.68	0.90	1.12
500	0.81	0.64	0.58	0.94	1.09
300	0.58	1.20	-0.79	1.40	1.32
200	-0.28	0.40	-1.11	-0.10	0.60
150	-0.48	-0.40	-1.06	-0.45	0.34
100	-0.61	-0.43	-0.80	-0.85	-0.40
70	-0.67	-0.15	-0.86	-1.04	-0.69
50	-0.59	-0.20	-0.70	-0.85	-0.72
30	-0.39	-0.15	-0.32	-0.56	-0.46
20	-0.04	-0.06	0.17	-0.09	-0.15
10	0.59	0.03	1.09	0.77	0.86
<b>Hohenpeissenberg (1969 - 1988)</b>					
Surface	3.03	2.72	2.58	3.29	3.46
700	2.13	2.14	1.98	1.94	2.46
500	2.34	2.69	1.82	2.17	2.89
300	2.03	2.91	1.20	1.95	2.13
200	0.10	0.23	-0.33	0.60	0.35
150	-0.09	-0.41	-0.62	1.08	0.12
100	-0.39	-0.84	-0.20	-0.01	-1.04
70	-0.67	-0.81	-0.87	-0.14	-0.91
50	-0.61	-0.75	-0.74	-0.48	-0.58
30	-0.32	-0.48	-0.30	-0.19	-0.32
20	-0.23	-0.53	-0.13	0.00	-0.29
10	-0.51	-0.47	-0.45	-0.66	-0.40
<b>Payerne (1969 - 1988)</b>					
Surface	--	--	--	--	--
700	1.05	1.03	1.22	0.94	0.98
500	1.17	1.30	1.44	0.95	1.02
300	0.70	1.03	0.58	0.53	0.65
200	-0.09	0.11	-0.29	-0.25	0.13
150	-0.29	-0.24	-0.54	-0.25	-0.09
100	-0.61	-0.76	-0.75	-0.44	-0.53
70	-0.79	-0.76	-0.98	-0.71	-0.75
50	-0.73	-0.61	-0.91	-0.73	-0.72
30	-0.43	-0.49	-0.44	-0.44	0.42
20	-0.22	-0.40	-0.18	-0.16	-0.17
10	0.11	-0.20	-0.10	0.27	-0.19

butions particularly for the stratosphere. The positive trends in the troposphere found over Hohenpeissenberg are very strong, ~2%/yr to 3%/yr, and occur during all seasons. At Payerne, the positive trends are ~1%/yr and are barely significant. In both cases the maximum positive trends are found at ~500 mb during all seasons. In the stratosphere the strongest negative trends (-0.9%/yr) occur during spring and fall over Hohenpeissenberg and during spring over Payerne.

The overall patterns of the ozone trends at mid-latitude and subpolar-latitude stations discussed above are generally consistent, but details of the seasonal variations show differences, particularly in the troposphere, that certainly reflect local influences.

Individual seasonal trend values are listed for the Canadian composite, Hohenpeissenberg, and Payerne in Table 1. The calculated trends for the Hohenpeissenberg and Payerne shown in Table 1 cover the same 20-year period (1969-1988) for comparison purposes. Note that there are no trend values given for the surface at Payerne. For the Canadian composite data, the largest positive annual trend is at the surface (1.2%/yr), and the largest negative annual trend (-0.7%/yr) is found in the stratosphere at 70 mb. The positive trends in the troposphere are significantly different from zero (2σ). In the stratosphere the negative trends are statistically significant (2σ) at levels from 30 to 100 mb. The largest positive trends occur during winter, summer, and fall and are generally found in mid-troposphere. Al-

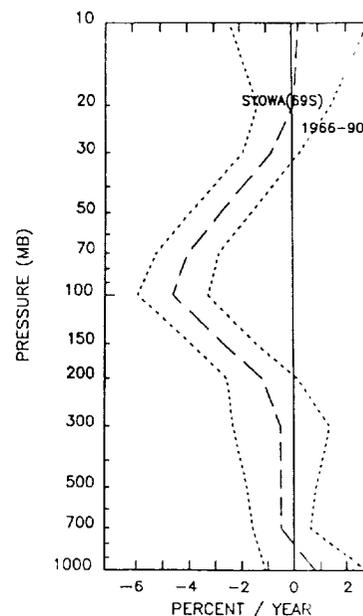


Fig. 3. Quarter 4 ozone trends for Syowa (69°S), 1966-90.

though there are large positive trends indicated at the surface, they are frequently not statistically significant. Significant, negative trends in the stratosphere (-0.7%/yr to -1%/yr) occur during spring, summer, and fall but not during winter. The trends are largest during spring, and the pressure level for the large negative trends decreases from 200 mb in spring to 50 mb in fall.

At Hohenpeissenberg the strong positive trends in the troposphere are present during all seasons of the year. In mid-stratosphere, however, the negative trend is just statistically significant (2σ) at 50-70 mb during spring and fall. The maximum positive trends at Payerne occur at ~500 mb and also do not show much seasonal variation. The negative trends in the stratosphere (-0.7%/yr to -0.9%/yr) are again found at ~50-70 mb. The seasonal variation of crossover level from positive to negative trends for Hohenpeissenberg and Payerne is similar to that found for the Canadian composite. This level occurs at the highest pressure generally during spring (or winter-spring as is the case for Hohenpeissenberg) and at the lowest pressure during fall (or summer-fall at Hohenpeissenberg). It is clear that the long-term patterns of ozone variations at these stations in mid-latitudes to polar latitudes of the Northern Hemisphere respond to different photochemical and dynamic influences in the troposphere and lower stratosphere (e.g., Liu et al., 1987; Austin et al., 1991).

Only one station in the Southern Hemisphere (Syowa, 69°S) has a sufficiently long period of available ozone-sonde observations to determine tropospheric and lower stratospheric trends. Even so the data set is adequate for only the spring season (September, October, and November) when the Antarctic ozone hole has become progressively more intense. The vertical distribution of the ozone trend at Syowa is shown in Fig. 3. The

trend in the troposphere is slightly negative but is not significantly different from zero, even at the  $1\sigma$  level. It is important to note that the lack of a tropospheric ozone trend in the Antarctic region is in contrast to the strong positive trend in the troposphere found for Northern Hemisphere mid- and polar-latitude stations. This is an indication of the minimum effect of low tropospheric photochemical influences in the Southern Hemisphere at high latitudes far removed from precursor pollutant sources. In the stratosphere, however, the negative trend from 40 to 200 mb is everywhere statistically significant ( $>2\sigma$ ) and at 100 mb reaches a value of  $\sim -5\%/yr$ .

## CONCLUSIONS

Analysis of the long-term ozone data at mid- to polar-latitude stations of the Northern Hemisphere shows a distinct pattern of increasing ozone in mid-troposphere and decreasing ozone in the lower to mid-stratosphere. This pattern occurs during all seasons, but the stratospheric decreases seem to be larger and generally more significant during the spring. The crossover from positive to negative ozone trends occurs in the upper tropospheric and lower stratospheric regions at the time of the largest exchange between the two regions.

What are the associated atmospheric effects of these trends? The major ozone decreases in the stratosphere occur at levels of highest ozone concentrations. Thus the negative stratospheric trends are reflected in negative trends of total ozone. Also, it has been shown that the observed stratospheric ozone trends are associated with observed and calculated negative trends in temperature above  $\sim 15$  km (e.g., Miller et al., 1992). It is clear that a total ozone loss will increase the transmittance of UV and visible irradiance to the surface and lower troposphere. However, the optical depth of ozone in the IR ( $9.6 \mu\text{m}$ ) in the troposphere is relatively small as compared with the optical depth at the ozone peak. Thus, surface radiation to space would normally increase as the ozone peak gets thinner. On the other hand, if the lower tropospheric ozone concentration continues to increase, the downward irradiance in the IR window is increased, leading to a contribution to a greenhouse-type warming. Simple 1-D radiative equilibrium models using typical observed vertical distributions of ozone variations are best suited to test the effectiveness of these variations as potential greenhouse contributions.

## ACKNOWLEDGMENTS

I thank Larry Morrison who made available corrected ozonesonde data archived by the World Ozone Data Center. I would also like to acknowledge the programming assistance given by Mary Eberle and Delbert Wagner. Support for this study was provided through the Upper Atmosphere Research Program of NASA under Contract No. NAS5-27263.

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